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TITLE: WORKING WITH SOME OF THE NEWER HIGH-Z SCINTILLATORS

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Working with some of the newer high-Z scintillators

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ABSTRACT

We have performed simple laboratory studies of several of the newer high-Z inorganic scintillators, with a primary goal of evaluating their relative merits as sensors for space-borne instruments. It is appropriate to consider using one of these materials when high light output can be exchanged for some other beneficial property, such as higher stopping power (bismuth germanate), superior resistance to radiation damage (barium fluoride), or fast decay time (barium fluoride and pure cesium iodide). Our work has revealed or confirmed other important characteristics—both favorable and unfavorable—of these newer scintillators. For example, the temperature dependence and particle species dependence of the fast and slow scintillation output of barium fluoride and pure cesium iodide will be described.

1. INTRODUCTION

In recent years, several new, high-Z, inorganic scintillators have become available. These include bismuth germanate ($Bi_4Ge_3O_{12}$ or BGO), barium fluoride (BaF_2 or BF), pure cesium iodide (CsI), and cerium fluoride (CeF_3 or CF). While none of these threaten to drive activated sodium iodide or cesium iodide off the shelves, all have properties that may be advantageous in certain applications. For example, we at Los Alamos often design and build space instruments that must be as compact as possible and must operate in a hostile radiation environment. Consequently, properties such as high stopping power, radiation damage resistance, and fast time constants are more valuable to us than the highest possible light output or superior energy resolution. Generally speaking, using one of the new scintillators means sacrificing (at least) the last two qualities in return for something else. In this paper we will discuss the advantages and disadvantages of BGO, BF, CsI, and CF. Other work will be cited, but we will draw primarily upon our own experience. Because of our requirements at Los Alamos, properties advantageous for modest space instrumentation will be emphasized.

2. EXPERIMENTAL TECHNIQUE

Since our intent was to obtain comparisons rather than absolutes, we stressed simplicity and reproducibility of test conditions. No attempt was made to obtain the best possible energy resolution or the fastest possible pulse rise times. The only optimization we did was when using BF with a glass-window photomultiplier. For most of the data, no optical coupling material was used, and aluminum foil was the reflector. The photomultipliers used were an EMI 9054QA (2-in.-diam, quartz window) and an EMI D509NA (1.5-in.-diam, compact, rugged). The total scintillation light was measured at the PMT anode using a charge preamplifier with a long integration time constant. A fast preamp (10 ns) measured the signal at the last dynode. Either the fast or total signal was routed through an Ortec 450 amplifier to a Canberra 35-Plus MCA. For a few of our samples, measurements were taken over a temperature range of −15 to +45 deg. C, and after exposure to high levels of radiation from a Co60 source. We also measured the response of the scintillators to neutrons. Some of our results, particularly for BGO, were obtained from other tests conducted over the past several years.

3. RESULTS

(a) Bismuth germanate

BGO has been recognized as a scintillator since 1975[1], which nearly disqualifies it from being called "newer." It is known for having the highest stopping power, low afterglow, and good physical properties. It has been the subject of numerous publications, and has been used by us in space instrumentation for several years. Accordingly, we will limit our discussion of BGO to the mention of a few miscellaneous points.

(1) Low afterglow cannot be considered a "given." We have observed increased noise levels in some crystals exposed to modest amounts of room light or gamma-radiation. Such samples also exhibit severe gain degradation vs radiation exposure.

(2) Radiation damage effects can become appreciable at about 100 rem[2]. Some samples spontaneously recover on time scales of hours or less, while others do not. We have even observed regions within a single crystal to differ from one another in this regard. Heating to 60 - 80 deg. C for a few hours usually heals any long-term radiation damage, but the temperature required is sample-dependent. The sample that we used in our recent testing showed a 17% gain reduction after an overnight exposure to 1000 rem, and recovery was nearly complete after 24 hours. Based on prior experience, this is excellent performance for a BGO crystal.

(3) Consistently accurate results in tests designed to optimize BGO-based detectors are difficult to obtain. Reasons for this include a high sensitivity of the gain to temperature variations (about 1% per deg. C), surface contamination (unpainted crystals must be cleaned often if they are handled or if optical coupling greases are used), and prior light exposure.

(b) Barium Fluoride

BF is similar in stopping power and physical properties to CsI, and its total light output is comparable to BGO. However, it has several well known distinguishing characteristics[3]. First, about 10% of its light output is in an extremely fast (<1 ns) component, with the actual percentage of fast component being significantly lower for heavily ionizing particles than for electrons. Second, the scintillation output is primarily ultraviolet; the fast component has the shortest distribution of wavelengths, with the output peaking near 220 nm. Some fast output is obtainable with glass-window PMTs using a fast wavelength shifter such as p-terphenyl, but a quartz window is required for best performance. Even with a quartz window, p-terphenyl can increase the fast-component response by shifting the spectrum to a region of higher PMT quantum efficiency. Third, many (but not all) BF crystals have a very high intrinsic background caused by radium contamination. Amazingly, only a few parts in 10^{10} of radium can give rise to thousands of counts per second in a 2" x 2" crystal. However, since this background is essentially all alpha particles (which produce little fast component), it can be suppressed using a fast/slow coincidence technique. In fact, the alphas could be used as a gatable inflight calibration, and a fast/slow coincidence requirement could be used to eliminate or separate detector counts caused by any heavily ionizing particles. When contemplating any such scheme, however, it must be borne in mind that the amplitude of the fast component is only ~3% of NaI.

Thermal testing of BF crystals procured from Bicron Corp. showed that for a p-terphenyl coated crystal the total light output had a negative temperature coefficient in excess of 1% per °C at room temperature. The temperature dependence was twice this value at 40° C, and virtually disappeared below 5° C. The fast component seemed independent of temperature. A bare BF crystal from the same lot appeared to behave identically, within the experimental uncertainties.

Bare and coated BF crystals were subjected to the same 1000 rem overnight dose as was the previously mentioned BGO crystal. Neither BF crystal showed any gain change, but both showed a small amount of noise below about 40 keV equivalent. Initially, the noise level was about 100 counts/sec, and it disappeared with a time constant of a few minutes. Its origin and detailed behavior need further investigation.

(c) Pure cesium iodide

The scintillation properties of CsI are unlike those of either CsI(Tl) or CsI(Na)[4]. The light output is slightly less than that of BF or BGO, and it consists mainly of a fairly fast (~10 ns) component. The spectrum is quite blue, peaking at 305 nm, but this is not disastrous for a glass-window PMT. We have procured samples from Harshaw and Optovac. At 25°C all of our samples emitted about half of their light in a slow component ($\gtrsim$5 microsec) when irradiated with gamma rays. The slow fraction increased monotonically and dramatically from only 0.12 at -5° C to about 0.67 at 45° C. The total scintillation output increased by 20%, or 0.4% per degree C, over this temperature range. Use of a slightly shorter preamplifier time constant could in principle result in zero net gain change with temperature.

The example of BF prompted us to measure the fast fraction for alpha particles as well as for gamma-rays. We found that, in direct contrast to BF, the alpha particle response was essentially 100% fast component. CsI (Tl) apparently exhibits a similar, but less striking behavior.[5]

We irradiated three CsI samples along with the BGO and BF. At first, our results for these crystals seemed confusing and contradictory. Eventually, we realized that the samples had such tremendous afterglow that our PMT gain was completely unstable. One of the samples, after an additional 300 rem of radiation, visibly glowed in the dark. Needless to say, this could be a serious problem in many applications. The glow disappeared with a time constant of minutes, although our worst sample was still significantly noisy after 24 hours. The (prompt) light output of the crystals always recovered. It is possible that the output in fact never varied; we could not measure it accurately in the presence of the afterglow.

At this point in time, we do not know if the "entertaining" behaviour we have observed from CsI is inherent to the pure material, or if a reduction of impurity levels will result in a more docile substance.

(d) Cerium fluoride

We have ordered CF crystals from Optovac, the only present supplier that we know of, but as of this writing the samples had not yet been delivered. This newly recognized scintillator is reputed to have about 0.5 times the light output of BGO, a 27 ns time constant, and a wavelength of 340 nm at the peak of the emission.[6] With a density of 6.16 gm/cm**2 and a Z of 58 for Cerium, its stopping power will be somewhat better than CsI or BF. Its physical properties are good, being similar to other commonly used fluorides. Hopefully, our samples will arrive in time to be discussed at the meeting.

(e) Other observations

Some of our results did not warrant individual mention in Sections (a) through (d) above because of insufficient differences between the materials. For example, the tested scintillators all responded similarly to a 10^4 s^{-1} Americium/Boron neutron source, yielding 2 to 4 x 10^{-4} counts n^{-1} gm^{-1} above 1 MeV. CsI would be the poorest choice if low neutron response were crucial. In the "real world" of procurement, we found all the materials except CF to be readily available from at least two sources. Finally, the cost of all 4 scintillators starts at about $2 gm^{-1} and ranges upward according to specifications.

4. ACKNOWLEDGMENTS

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